

### **REMARKS/ARGUMENTS**

Upon entry of the above amendments, claims 20-27 will be pending. Claim 20 corresponds substantially to previous claim 11, with the addition of the subject matter of claim 16 and with the further addition of subject matter from claim 19. Claims 21-27 substantially correspond to previous claims 12-15 and 17-19, respectively.

The Office Action had rejected claims 11-19, under 35 U.S.C. 102(b), as anticipated by, or under 35 U.S.C. 103(a), as obvious over Quinlan *et al*, US 4,812,300 (the '300 Patent).

Applicants respectfully disagree and, therefore, reconsideration and allowance is respectfully requested.

As noted from a review of the pending claims, the process under consideration relates to the oxidation of ammonia by reacting ammonia and air in the presence of an oxidation catalyst. The oxidation catalyst is an at least ternary cobalt mixed oxide phase catalyst system containing both variable valency metal (Vv) and non-variable valency metal (Vn).

The process according to the pending claims provides improved activity and selectivity in comparison to binary catalyst systems and other ternary catalyst systems.

The '300 Patent does not anticipate or make obvious the claimed process.

According to the '300 Patent, ammonia oxidation may be carried out using a mixed oxide phase (perovskite) catalyst. The perovskite catalyst has formula  $ABO_3$  wherein A is a cation of an element selected from among alkali, alkaline earth, rare earth, lanthanide, actinide metals or a mixture of these metals having a relatively large ionic radius (*see, e.g.*, col. 4, lines 61-68; col. 5, lines 1-12 and 51-54). Within this range, particularly preferred cations A are mixtures of lanthanum with alkaline earth metals, especially strontium (*see, e.g.*, col. 5, lines 54-60).

The B metal ion is a cation of an element selected from Groups IB, IVB, VB, VIB, VIIB or VIII of the Periodic Table (*e.g.* col. 5, lines 64-66). The Groups VIIB and VIII metals are preferred (*e.g.*, col. 5, lines 67-68).

Although the B cation may be cobalt (col. 6, lines 1-3), the A cation combinations described in the '300 Patent do not specifically include a variable valency element Vv selected from cerium and praseodymium combined with a non-variable valency element Vn selected from non-variable valency rare earths and yttrium. Indeed, the preferred A cations in

Quinlan et al are lanthanum, alkaline earth cations or mixtures thereof, none of which are variable valency cations (*see, e.g.*, col. 5, lines 54-56 and col. 7, lines 1-9).

Therefore, the '300 Patent does not disclose an ammonia oxidation process using a catalyst comprising oxides of (a) at least one element Vv selected from cerium and praseodymium and at least one element Vn selected from non-variable valency rare earths and yttrium, and (b) cobalt, with at least some of the cobalt and element Vv and element Vn oxides present as a mixed oxide phase.

Accordingly, claim 20 is novel over the '300 Patent. Since claims 21-27 are dependant upon claim 20, these claims are also novel over the '300 Patent for at least the same reasons.

Furthermore, for at least the following reasons, the process as set forth in claims 20-27 would not have been obvious in view of the '300 Patent.

The only examples of ternary perovskite oxides with two A-site cations considered in the '300 Patent are alkaline earth-doped catalysts of formula  $A_{1-x}A'_xBO_3$  where A is any trivalent lanthanide cation, A' is divalent (i.e.  $2^+$ ) calcium, strontium or barium, particularly strontium, x is 0-1 and B is any transition metal having polyvalent ( $3^+$ ) and ( $4^+$ ) oxidation states (see column 7, lines 1-9). The only catalysts tested of this formula were strontium-doped lanthanum cobaltate and strontium-doped lanthanum manganate (Example 5 Table 1) in which the selectivity for NO was only slightly affected by the addition of Sr ( $2^+$ ) (column 9, lines 42-44).

This disclosure would not have rendered obvious an A-site dopant cation which is a variable valency element Vv, cerium (Ce) or praseodymium (Pr). Both cerium and praseodymium are termed variable valency because they may adopt a ( $3^+$ ) or ( $4^+$ ) oxidation state within the mixed oxide phase. Typically they may be included within the mixed oxide phase in their ( $4^+$ ) oxidation state. The presence of these variable valency elements will change the electronic properties of the oxide system and therefore effects the catalyst stability, activity and selectivity.

Alkaline earth elements such as strontium, because they are divalent, substitute the A-cation (e.g. lanthanum cation) to create an oxygen vacancy in the perovskite structure. Cerium or praseodymium, because they are tetravalent, do not and are believed to either lead to the formation of a vacancy on the Vn cation-site, a reduction in the oxidation state of the

cobalt or the filling of oxygen lattice vacancies which exist in the  $VnCoO_3$  structure. Therefore mechanistically, doping the mixed oxide phase with cerium or praseodymium results in different effects than would be observed with alkaline earth doping.

The Applicants have found that addition of even small amounts of cerium into the A-site of lanthanum cobaltate produces an ammonia oxidation catalyst with higher selectivity to NO than the un-doped catalyst. This is illustrated in Example 1 of the present application where a  $La_{0.75}Ce_{0.25}CoO_3$  catalyst provided higher selectivity than a  $LaCoO_3$  catalyst across a range of reaction temperatures.

In addition, further experiments on Sr and Ce-doped  $LaCoO_3$  have demonstrated the improved efficiency of the catalysts as defined herein when compared to catalysts within the scope of the '300 Patent. Catalysts having the composition  $La_{0.98}Sr_{0.02}CoO_3$  and  $La_{0.98}Ce_{0.02}CoO_3$  were prepared by the precipitation method described in Example 2 with final calcination at 900 °C for 6 hours. An un-doped  $LaCoO_3$  was prepared for comparison. The catalysts were then tested for their efficiency (i.e. the percentage of the ammonia converted to NO and  $NO_2$ ) in a lab-scale ammonia oxidation reactor. The catalysts were tested twice under the same conditions.

About 60 g of a pelleted catalyst was placed, as a 50 mm bed supported on a stainless steel gauze, in a stainless steel tubular reactor of 28 mm i.d. A mixture of ammonia (10-10.5% by volume) and air was preheated to about 180 °C and passed through the reactor. The air was fed at 2 m<sup>3</sup>/hour. The exothermic oxidation reaction taking place on the catalyst raised the gas temperatures to 850-900 °C. The resulting product gas mixture was analysed by mass spectroscopy and the efficiency calculated by taking the average ammonia conversion to NO/ $NO_2$  over a two hour period, measured after steady state conditions had been established. The reaction was stopped by switching off the ammonia and cooling the catalyst under air. The process was then repeated. A small improvement in efficiency is

normally observed for the second run due to catalyst conditioning. The results obtained are shown in the following table:

Catalyst	Run	Efficiency	Average
$\text{La}_{0.98}\text{Ce}_{0.02}\text{CoO}_3$	1	88.4	90.7
	2	93.0	
$\text{La}_{0.98}\text{Sr}_{0.02}\text{CoO}_3$	1	87.1	88.1
	2	89.1	
$\text{LaCoO}_3$	1	78.3	83.6
	2	88.8	

Taking the first, second or average efficiency figure, the results demonstrate that the Ce-doped catalysts give higher efficiency results than the Sr- or un-doped  $\text{LaCoO}_3$ .

It further submitted that the '300 Patent teaches away from the present invention. In a primary embodiment of the reference, the A and B cations are selected with the proviso that A or B are not the combination of cerium and cobalt or of lanthanum and cobalt (column 5, lines 61-63). As such, the practitioner would be led away from combinations which are present in the process according to the present invention. This is still further evidence that it would not have been obvious to provide a mixed oxide phase catalyst such as  $\text{LaCeCoO}_3$ , or any other catalyst within the scope of the pending claims.

Accordingly, reconsideration and withdrawal of the rejection, applied to claims 11-19, is respectfully requested.

For completeness, Applicants have also considered the disclosure of the cited but not applied, patent to Hughes *et al*, U.S. 3,888,792 (the '792 Patent).

The '792 Patent describes oxidation catalysts in the form of shaped bodies comprising mixtures of cobalt (II, III) oxide ( $\text{Co}_3\text{O}_4$ ) with oxides of scandium, yttrium, and the lanthanide elements.

Cobalt (II, III) oxide may be mixed with oxides of scandium, yttrium, and the lanthanide elements, or a mixture of at least two of these oxides (column 1, line 57). Only binary mixtures are explicitly described, see, *e.g.*, the Examples.

The '792 Patent does not disclose that the cobalt catalyst should be part of a mixed oxide phase, *e.g.*, a perovskite. The cobalt component of the shaped bodies is referred to only as "active cobalt (II, III) oxide", *i.e.*,  $\text{Co}_3\text{O}_4$ , *e.g.*, at col. 1, lines 51-52, col. 2, lines 4, 56 and 59. Indeed, the preferred catalyst system appears to be ceria-promoted cobalt (II, III) oxide

(Examples II, III & IV) but because of their relative ionic radii, cerium cannot form a stable mixed oxide phase with cobalt.

A further indication that the cobalt does not comprise part of a mixed oxide phase may be found at column 2, lines 54-58 where it is stated that the temperature at which the composition is heated when making the shaped catalyst bodies should be kept below that at which the  $\text{Co}_3\text{O}_4$  is converted to less active cobalt (II) oxide,  $\text{CoO}$ . If the cobalt was a component of a mixed oxide phase Applicants believe that it would not have the Hughes temperature limitation in respect of its preparation (600-850 °C, column 2, line 49).

The object of the '792 Patent appears to be to overcome the mechanical weakness of  $\text{Co}_3\text{O}_4$ -based catalysts by combining  $\text{Co}_3\text{O}_4$  with promoters having adequate mechanical strength for use in industrial processes (see, *e.g.*, col. 1, lines 40-44). The ammonia oxidation processes are operated at temperatures in the range 650-800 °C (col. 3, line 5), with the Examples employing reaction temperatures of only 650 °C and 700 °C. These, temperatures are below that at which the thermal degradation of the active  $\text{Co}_3\text{O}_4$  to less active  $\text{CoO}$  commences (about 850 °C, see, *e.g.*, col. 2, lines 54-59) and are below those typically used in modern ammonia oxidation processes (*e.g.* 850-950 °C).

The '792 Patent would not have made obvious the means to overcome the thermal instability of the  $\text{Co}_3\text{O}_4$ -based catalysts by forming a mixed oxide phase let alone a mixed oxide phase comprising at least one element Vv selected from cerium and praseodymium and at least one element Vn selected from non-variable valency rare earths and yttrium, and cobalt.


Accordingly, Applicants concur in the non-reliance on the '792 Patent as either anticipating or making obvious, the present invention.

Therefore, all objections and rejections having been addressed, it is respectfully submitted that the present application is in a condition for allowance and a Notice to that effect is earnestly solicited.

Should any issues remain unresolved, the Examiner is encouraged to contact the undersigned attorney for Applicants at the telephone number indicated below in order to expeditiously resolve any remaining issues.

Respectfully submitted,

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